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Application of radon and radium isotopes to groundwater flow dynamics: An example from the Dead Sea

Yael Kiro [a,b,*], Yishai Weinstein [c], Abraham Starinsky [a], Yoseph Yechieli [d,e]

[a] Institute of Earth Sciences, The Hebrew University, Edmond J. Safra Campus, Givat Ram, Jerusalem 91904, Israel
[b] Lamont-Doherty Earth Observatory, Columbia University, USA
[c] Bar-Ilan University, Department of Geography and Environment, Ramat-Gan 52900, Israel
[d] Geological Survey of Israel, 30 Malkhe Israel Street, Jerusalem 95501, Israel
[e] Department of Hydrology & Microbiology, Zuckerberg Institute for Water Research, Ben-Gurion University of the Negev, Midreshet Ben-Gurion 84990, Israel

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ABSTRACT

This study presents the behavior of radon and radium isotopes and their application to groundwater age and flow dynamics. The research was conducted in the complex Dead Sea groundwater system, which includes a large variety of sediments, groundwater salinities, flow mechanisms and groundwater ages. Groundwater around the Dead Sea contains high activities of radon (up to tens of thousands dpm/L) and radium (up to hundreds dpm/L). Adsorption of radium, which is partially salinity controlled, is an important source of unsupported ^{222}Rn, which is used for estimating the adsorption partition coefficient of radium. In addition to salinity, the concentration of Mn and Fe oxides and aquifer heterogeneity are important factors controlling the adsorption partition coefficient. The different nature of the rocks on both sides of the Dead Sea transform, with lower Th/U ratios in the carbonate rocks on the western catchment of the Dead Sea compared to higher ratios in the sandstone aquifer east of the Dead Sea, is reflected in a higher ^{228}Ra/^{226}Ra activity ratio in the eastern compared with the western groundwaters (averages of 0.76 and 0.15, respectively). The different groundwater groups around the Dead Sea contain secular or non-secular equilibrium ratios, which depend on the age of the groundwater (the time since the groundwater entered the aquifer) or whether the groundwater system is in a steady state (the age of the groundwater system). Young groundwater, such as the Dead Sea water that circulates in the aquifer or freshwater springs, is depleted in the long-lived radium isotopes compared to the short-lived isotopes, whereas old groundwater contains relatively high activity of ^{226}Ra (~500 dpm/L) and the radium activity ratios are close to secular equilibrium. The common secular equilibrium ratios between all four radium isotopes in the Dead Sea groundwaters suggest that many of the groundwater flow paths did not change significantly during the past 8000 years.

1. Introduction

Groundwater concentrations of radionuclides have been shown to be significantly affected by water–rock interaction (e.g. Rama and Moore, 1984; Davidson and Dickson, 1986; Tricca et al., 2001). In particular, the activities of radon and radium isotopes are usually high in groundwater as a result of the decay of their parent nuclides in the host aquifer solids (e.g. Krishnaswami et al., 1982). The concentrations of these radionuclides in groundwater are determined by the concentrations of U and Th in the aquifer rock, the travel time of the groundwater, recoil, dissolution and the adsorption–desorption processes (e.g. Davidson and Dickson, 1986; Tricca et al., 2001). However, the use of radon and radium isotopes in order to understand flow dynamics and estimate groundwater age and velocity has hardly been examined.

The objective of this study was to use the Dead Sea hydrological system as a natural field lab for studying the factors controlling radon and radium isotopes in different settings of groundwater and to use these isotopes to characterize water ages and dynamics. The Dead Sea hydrological system provides the opportunity to gain a comprehensive dataset of radon and radium isotopes of a diverse environment with different rock types, a large salinity range and different groundwater flow mechanisms and ages.

1.1. The behavior of radon and radium isotopes in groundwater

There are four naturally occurring radium isotopes (the 'radium quartet'), with half-lives ranging from 3.7 days to 1600 years. Two of them, ^{226}Ra and ^{223}Ra, are the radioactive progeny of uranium isotopes (^{238}U and ^{235}U, respectively), while ^{228}Ra and ^{224}Ra are the decay products of ^{232}Th. Three of the isotopes (^{226}Ra, ^{224}Ra and ^{223}Ra) disintegrate into radon isotopes (^{222}Rn, ^{220}Rn and ^{219}Rn, respectively), while ^{228}Ra decays to ^{224}Ra via ^{228}Th (and the short-lived ^{228}Ac).

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* Corresponding author at: Institute of Earth Sciences, The Hebrew University, Edmond J. Safra Campus, Givat Ram, Jerusalem 91904, Israel.
E-mail address: yael.kiro@mail.huji.ac.il (Y. Kiro).

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Several processes are responsible for radium input to the groundwater: a) alpha recoil by the decay of the parent nuclide (Th) in the aquifer solids; b) desorption from the solid surface; c) dissolution; and d) decay of the parent nuclide in the solution. Radium sinks include a) radioactive decay, b) adsorption (Krishnaswami et al., 1982) and c) co-precipitation (e.g. Kiro et al., 2012).

As for radon, which is a dissolved gas, the main sources include a) decay of dissolved ^{226}Ra; and b) alpha recoil by the decay of ^{226}Ra in the solid, while the main sinks are radioactive decay and loss to the pore space above groundwater level due to degassing. High activities of radon are common in groundwater and are typically ~5–10% of the total radon produced in the aquifer rock by the decay of ^{238}U and its daughters. This fraction is considered to be relatively large (Tricca et al., 2001; Porcelli and Swarzenski, 2003), taking into account the expected contribution by recoil (Kigoshi, 1971). In the Dead Sea groundwater, radon activities are much higher, and they often even exceed the activities produced in the rock (>100%, Kronfeld et al., 1991; Moise et al., 2000). Several models regarding the phase of ^{226}Ra in the solid or the mechanism of ^{222}Rn release from the sediments explain such high activities of ^{222}Rn. Krishnaswami et al. (1982) interpreted the high activities of radon as the production by alpha recoil from the uranium (supported ^{226}Ra) in the rock, while Moise et al. (2000) attributed the high activities to the high content of adsorbed unsupported ^{226}Ra on the solids. Other proposed sources include the diffusion of radon from clays, preferential release of radon through nanopores (Rama and Moore, 1984), leaching of parent nuclides or high concentrations of ^{226}Ra in small grains or secondary phases (Tricca et al., 2001; Porcelli and Swarzenski, 2003).

In a closed water–rock system, the total concentration of radium and radon in the solids and water are supposed to reach secular equilibrium with the ^{238}U, ^{235}U and ^{232}Th isotopes. Radium is partitioned between the solid surface and the groundwater according to the adsorption partition coefficient (K), and in steady-state conditions the ^{226}Ra/^{223}Ra and ^{224}Ra/^{228}Ra ratios in the water should represent values around secular equilibrium (Davidson and Dickson, 1986). ^{224}Ra/^{228}Ra ratio in secular equilibrium is 1, while ^{226}Ra/^{223}Ra activity ratio is ~21 according to the natural abundances and the decay rates of ^{238}U and ^{235}U. Due to the effect of recoil, the number of alpha decay steps and the distribution of U and Th in different phases of the rocks, the ratios of ^{224}Ra/^{228}Ra in the water of such a system may vary between 0.67 and 1.5, while ^{226}Ra/^{223}Ra may reach twice the ^{238}U/^{235}U activity ratio and ^{226}Ra/^{228}Ra ratio may reach twice the U/Th ratio in the solid (Krishnaswami et al., 1982; Davidson and Dickson, 1986; Porcelli and Swarzenski, 2003). The deviation of these isotope activity ratios from secular equilibrium depends on the rate of water–rock processes, the age of the groundwater and groundwater flow paths and mechanisms (Davidson and Dickson, 1986; Porcelli and Swarzenski, 2003).

Although the applications of radium isotopes to groundwater hydrology was already suggested (e.g. Davidson and Dickson, 1986; Porcelli and Swarzenski, 2003), the main focus of these studies was on characterizing the behavior of radium and radon in groundwater, given the hydrological conditions, whereas only a few studies used these isotopes to actually calculate groundwater age or velocities (e.g. Krest and Harvey, 2003).

1.2. The Dead Sea hydrological and geochemical system

The Dead Sea is a terminal lake located in a deep pull-apart basin along the Dead Sea transform fault. The lake level has been continuously dropping from ~−390 m in the 1930s (Klein and Flohn, 1987) to −428 m at present (2014), reaching a rate of more than 1 m/year. This decline is the result of the increased utilization of fresh water in the northern part of the basin and the increased evaporation at the salt ponds of the Israel Dead Sea Works and the Jordanian Arab Potash Company. Altogether, these changes in water management resulted in a decrease of the average annual inflow to the lake from 1600 to 2000 million m^3 in the early 1900s (Neumann, 1958; Klein, 1998; Salameh and El-Naser, 1999) to a current 265–335 million m^3 (Lensky et al., 2005).

The composition of the Dead Sea water is Ca-chloridic with a very low Na/Cl ratio compared to normal ocean water (0.2 today vs. 0.86, respectively). This composition is the result of the evaporation of seawater that intruded the rift valley, followed by halite precipitation and dolomitization (Starinsky, 1974; Katz and Starinsky, 2008).

The present Dead Sea salinity and density are 340 g/L and 1.24 kg/L, respectively. It is extremely rich in radium relative to most natural water bodies. The ^{226}Ra activity in the Dead Sea (~147 dpm/L; Stiller and Chung, 1984; Kiro et al., 2012) is 1600 times that of Pacific surface water (~0.09 dpm/L) and 400 times that of Pacific deep water (~0.35 dpm/L, Broecker et al., 1967).

The present water sources of the Dead Sea are the Jordan River (~100 million m^3/year), fresh and brackish groundwater and brines discharging along the western shore (~100 million m^3/year) and perennial streams on the eastern catchment of the Dead Sea (~100 million m^3/year, Lensky et al., 2005; Salameh and El-Naser, 1999, Fig. 1). Almost all water sources of the Dead Sea are groundwater-based and therefore contain relatively high activity of radium. The main source of water in the western Dead Sea is the Ein Feshkha springs, which are brackish and discharge at a relatively constant salinity (~2 g Cl/L, ~60 million m^3/year). The Kane, Samar and Ein Gedi springs (Fig. 1) are of fresh water (<0.5 g Cl/L). These springs, as well as Ein Feshkha, are mainly fed by rain water infiltration in the highlands of the Judea Mountains. The difference in salinity is due to the mixing of the fresh meteoric groundwater with circulating Dead Sea water or residual brines. The Ein Qedem springs (Fig. 1) are thermal brines (~120 g Cl/L), with a relatively low discharge (~10 million m^3/year, The Hydrological Service of Israel). However, these brines are a major source of dissolved constituents in the lake due to their high salinity. The temperature of the thermal brines is around 45 °C, which indicates that the groundwater comes from a depth of at least 0.5 km (Gavrieli et al., 2001; Stern, 2010).

Unlike most of the western side, the eastern catchment of the Dead Sea is characterized by perennial streams, which originate from fresh to brackish springs, some of which are thermal (up to 65 °C), while just a small water volume derives from floods (Salameh, 1996). The main streams include Wadi Mujib, Wadi Hasa and Wadi Zarqa Ma'in (Fig. 1). At present, most of these water sources are dammed and utilized.

There are three main aquifers on the western side of the Dead Sea: the Lower Cretaceous sandstone Kurnub aquifer (~300 m thickness), which is isolated and has no direct contact with the other aquifers; the Upper Cretaceous carbonate Judea aquifer (~500 m thickness);and the Quaternary alluvial aquifer (usually ~50–100 m thickness; Arad and Michaeli, 1967; Yechieli et al., 1995). The alluvial aquifer is in direct hydraulic contact with the lake along most of its shoreline. It mainly consists of clastic sediments (mainly carbonates) deposited in fan deltas (gravel and sand) and of lacustrine sediments (e.g. clays, aragonite, gypsum and halite). The alluvial aquifer is bounded on its west by normal faults, which set Cretaceous carbonate rocks of the Judea Group against Quaternary alluvial and lacustrine sediments. The freshwater recharge of the alluvial aquifer is mainly through lateral flow from the Judea Group aquifer, which is replenished in the highlands 10–30 km to the west and by flashfloods. Direct recharge of this aquifer is negligible because of the arid climate in the Dead Sea region (precipitation of <100 mm/year). On the eastern side of the Dead Sea the groundwater aquifers are divided into three: the deep sandstone aquifer; the Upper Cretaceous carbonate aquifer; and the shallow aquifer. The sandstone aquifer contains two units, the Paleozoic Disi Group and the Jurassic–Lower Cretaceous Kurnub and Zerka groups, both which outcrop along the Dead Sea Rift valley (Salameh, 1996). Most groundwater discharges from the carbonate and the sandstone aquifers.

The hydraulic relationship between the Dead Sea and the alluvial aquifer is expressed in a relatively rapid groundwater level response